



Coupling of anodic and cathodic modification for increased power generation in microbial fuel cells

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H I G H L I G H T S

- Coupling effect of anodic and cathodic modification in single-chamber MFCs.
- Carbon mesh materials were modified by nitric acid or hydrazine hydrate to improve performance.
- Power density and CE after nitric acid modification of both electrodes enhanced by 38% and 32.2%.
- Chemical reduction method shortened accumulative time and enhanced power density.

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Carbon mesh (CM) materials are modified by nitric acid or hydrazine hydrate to test whether the performance of MFCs could be improved. The power densities of MFCs using nitric acid-treated anode (CM-NA) and hydrazine hydrate-treated anode (CM-HA) are improved by 24% ($811 \pm 24 \text{ mW m}^{-2}$) and 19% ($777 \pm 35 \text{ mW m}^{-2}$) as compared to the unmodified control ($655 \pm 7 \text{ mW m}^{-2}$). All MFCs using modified cathodes (CM-NC/Pt, CM-HC/Pt) also show higher performance in electrochemical response and power generation. The maximum power densities of reactors using CM-NC and CM-HC are respectively $811 \pm 29 \text{ mW m}^{-2}$ and $792 \pm 16 \text{ mW m}^{-2}$, which is 24% and 21% higher than the control. XPS and SEM results show that the performance improvement is related to the changes of surface functional groups and surface area. Further, the power densities with both anode and cathode modified by nitric acid (NN) and hydrazine hydrate (HH) are found to be increased by 38% ($905 \pm 15 \text{ mW m}^{-2}$) and 31% ($860 \pm 30 \text{ mW m}^{-2}$), respectively. CV measurements indicate that the electricigens have much higher activity. These results demonstrate that the power output of MFCs can be further increased through coupling of anodic and cathodic modification.

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1. Introduction

Microbial fuel cells (MFCs) are novel devices for direct bioelectricity generation from organic matters oxidation [1–3]. A MFCs reactor generally consists of an anode, a cathode, and sometimes a separator between the two electrodes. Biofilms on the anode oxidize various biodegradable substrates, and then electrons are transferred to the cathode through an external circuit. At the cathode, protons and electrons typically are catalyzed to react with oxygen, forming water [4,5]. The ability to generate power from wastewater makes MFCs a promising technology for wastewater treatment [6]. Although the power output has been enhanced in recent years, it is still far from utilizing it in practical applications

[7,8]. In order to make the process cost-effective, the power density needs to be further increased.

A large quantity of efforts have been made on electrodes to enhance energy production in MFCs. Carbon-based electrodes are perhaps the most commonly utilized because they are inexpensive and are readily attached by electrogenic bacteria. Chemical and physical modifications of the anodes are successful approaches for improving power production. The anode performance is related with its surface chemistry [9]. Cheng and Logan showed that power production was improved by 20% when a carbon cloth was treated using a high-temperature (700 °C) ammonia gas process, which was likely due to the increased amino groups on the anode surface [6]. The combination of heating and acid soaking treatment increased power from 1020 mW m^{-2} to 1370 mW m^{-2} , which was 34% larger than that of the untreated control [4]. Using activated carbon fiber felt anode after nitric acid pretreatment, the power

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output of MFCs improved by 58% reaching 2066 mW m⁻² compared to the control (1304 mW m⁻²) [10]. Zhou et al. developed a carbon mesh anode modification method using electrochemical oxidation in nitric acid electrolyte, and observed the maximum power density was 43% larger than the unmodified control [11]. The improvement of power generation using these modified methods was supposed to the changes of surface functional groups on the anode surface, which were good for the attachment and growth of electrogenic bacteria on the anodes.

On the other hand, the cathodic overpotential is also a limiting drawback that impacts power production, due to the reaction kinetics of oxygen reduction rate [12,13]. Considerable attention has been paid to the cathode modification with low cost catalysts to reduce the activation energy, including MnO₂ [14], Mn₂O₃ [15,16], Fe₂O₃ [15], polypyrrole (Ppy) [17], iron phthalocyanine (FePc) [18,19], cobalt and iron–copper phthalocyanine (CoPc and FeCuPc) [18], cobalt tetramethoxyphenylporphyrin (CoTMPP) [18,19], stainless steel alloy and NiMo [20]. However, Pt is still one of the most effective catalysts in the current application. Moreover, growing literature suggest that the power output of MFCs improves along with the increase of cathode specific surface area. Nitric acid and thermal activation of high specific surface area graphite granules in the cathodic compartment of a dual-chamber MFC led to a higher open circuit voltage [21]. The power density increased from 67 mW m⁻² to 315 mW m⁻² when plain carbon paper cathode was replaced with high specific surface area activated carbon felt cathode [22]. Cheng and Logan demonstrated that the volumetric power density had a linear relationship with the cathode specific surface area [23]. Wang et al. developed carbon nanotube (CNT) modified 3-D air-cathodes which showed higher performance in electrochemical response and power generation [24].

While these methods all result in power improvement, the development of MFCs for actual applications will require less expensive and simpler modification methods for higher power generation. Surface modifications could introduce some hydrophilic groups like amino compounds to carbon surfaces. Previous researches focused on anode or cathode modification individually. However, to the best of our knowledge, no report has addressed the enhancement of MFCs performance when both anode and cathode are modified. In this study, we presented the coupled function of anode and cathode modification on the MFCs performance for the first time using nitric acid or hydrazine hydrate pretreated electrode material as anode and cathode. And it verified that the power output of MFCs could be considerably enhanced through this coupling compared to their individual modifications either on anode or cathode.

2. Materials and methods

2.1. Anodes and cathodes preparation

Carbon mesh (Jilin Carbon Plant, China) was first cleaned by soaking in pure acetone overnight and then washed by deionized water for future use and modification. This pretreated carbon meshes were denoted as CM-CA. For nitric acid treatment, the CM-CA samples were soaked in concentrated nitric acid for 5 h, and then were washed by deionized water until the pH was about 7.0. Then, they were dried in 120 °C for 2 h (CM-NA) for future use [10]. For hydrazine hydrate treatment, the CM-CA samples were soaked in ethanol solution with 30% hydrazine hydrate, heating at 60 °C and refluxing for 6 h. Subsequently, they were dried in oven at 105 °C for further use (CM-HA).

Cathodes were made of pretreated carbon mesh containing a Pt catalyst (0.2 mg cm⁻²) on the water-facing side, with one carbon base layer and four PTFE diffusion layers on the air-facing side, as described by Cheng et al. [25]. They were denoted by the

unmodified cathode (CM-CC/Pt), the nitric acid-treated cathode (CM-NC/Pt), the hydrazine hydrate-treated cathode (CM-HC/Pt), respectively. And the cathodes without Pt were labeled as CM-CC, CM-NC, CM-HC, respectively.

2.2. MFC configuration and operation

Membrane free single chamber air-cathode MFCs (14 mL volume) were constructed as previous described [11,26]. Various surface modified electrodes were used to investigate the variation of power density. MFCs were inoculated with 20% anaerobic sludge (TEDA Sewage Treatment Plant, Tianjin), and a medium containing glucose (1 g L⁻¹) and a 50 mM phosphate buffer solution (PBS: NH₄Cl 0.31 g L⁻¹, KCl 0.13 g L⁻¹, NaH₂PO₄·2H₂O 3.32 g L⁻¹, Na₂HPO₄·12H₂O 10.36 g L⁻¹) [27] containing vitamins (5 mL L⁻¹) and minerals (12.5 mL L⁻¹) [28]. This solution was switched to a feed solution containing glucose (1.0 g L⁻¹) and PBS (50 mM, pH 7.0) when the similar output voltage produced over two consecutive cycles [11]. The feed solution was replaced when the voltage dropped below 50 mV. The external resistance was fixed at 1000 Ω and all tests were operated in duplicate in a fed-batch mode at 30 ± 0.5 °C in a temperature-controlled biochemical incubator.

2.3. Analysis methods

The voltages (V) across the fixed external resistor were measured every minute using a data acquisition system (PISO-813, ICP DAS Co., Ltd.) connected to a personal computer. Polarization curves were obtained by applying a different external resistance (*R*, from 10,000 Ω to 100 Ω) to the circuit and the maximum sustainable voltage was recorded for each resistance. Current density (*I*) was calculated from $I = V/R$, and normalized to the projected cathode surface area. Power density (*P*) was calculated using $P = IV$, and normalized to the projected cathode surface area [29]. The chemical oxygen demand (COD) was measured with the closed reflux spectrophotometric method on a commercial COD detector (HACH, DRB 200, DR/890 Colorimeter, USA). The Coulombic efficiency (CE) was calculated based on total COD removal over the cycle, as previously described [29,30].

Electrochemical measurements were carried out with CHI660D workstation (CH Instruments, Chenhua, Shanghai, China) connected to a personal computer. Linear sweep voltammetry (LSV) was used to measure the electrochemical performance of the cathodes. These tests were conducted in a three-electrode single-chamber reactor containing a working electrode (7 cm² projected area), a platinum wire counter electrode, and a SCE reference electrode [3]. Electrochemical activity of the anodic bacteria was measured by cyclic voltammogram (CV) in the same reactor with the anode as the working electrode, the cathode as the counter electrode, and SCE as the reference electrode. All experiments were performed in 50 mM PBS in the absence of nutrients at room temperature.

The surface morphologies of the anodes before incubation were examined by a field emission scanning electron microscope (FESEM, Hitachi, S-4700). The XPS measurements were carried out on X-ray photoelectron spectra (PHI 5000C ESCA System, Physical Electronics, USA) with Mg Kα radiation ($h\nu = 1253.6$ eV).

3. Results and discussion

3.1. Cathode modification

3.1.1. Electrochemical characterization of cathodes

The electrochemical performance of the cathodes can be observed by the decrease in current density during LSV testing

compared to the unmodified control in the absence of bacteria (Fig. 1). Among the cathodes without Pt catalyst coating, those modified electrodes (i.e., CM-HC and CM-NC) showed much larger current responses than the unmodified control (CM-CC). The CM-CC hardly showed any current response across the potential scan range, indicating that the carbon mesh itself did not catalyze oxygen reduction [24]. The higher specific surface area and the increase of electrochemically active groups were probably the reasons for the larger limiting current densities of modified cathodes without Pt, which were confirmed by SEM and XPS observations [5,24,31]. Clearly, the Pt catalyst significantly increased the current densities of all cathodes. The current densities of CM-CC/Pt, CM-HC/Pt, and CM-NC/Pt cathodes all increased substantially compared with their non-Pt counterparts and the CM-NC/Pt showed the best performance with the highest current response. These results illustrated that CM-NC/Pt cathode had excellent catalytic activity for the oxygen reduction reaction.

3.1.2. MFC performance

The modified air-cathodes were transferred to pre-acclimated single chamber MFCs after LSV tests. Based on the polarization and power density curves, the maximum power densities produced by different treatments were both larger than that of the control (CM-CC/Pt; $655 \pm 7 \text{ mW m}^{-2}$) (Fig. 2). Hydrazine hydrate treatment increased power density by 21%– $792 \pm 16 \text{ mW m}^{-2}$, and nitric acid treatment produced the power density of $811 \pm 29 \text{ mW m}^{-2}$, which was 24% larger than the control. The power production in MFCs tests were similar to the results observed during LSV tests.

3.2. Anode modification

3.2.1. Power generation using different surface modified anodes

MFCs were refueled every 24 h until the similar output voltage produced over two consecutive cycles. The start-up time of CM-NA was 113 h before reaching the first maximum power production, and that of CM-HA was only 67 h. The control MFC (CM-CA) required 122 h before reaching the first maximum power production (Fig. 3A). The time to achieve maximum repeatable voltages of the CM-NA and CM-HA was shortened by 7% and 45% as compared to that of the control.

Fig. 3B shows the polarization and power density curves of MFCs operating on different anodes. It could be observed that the

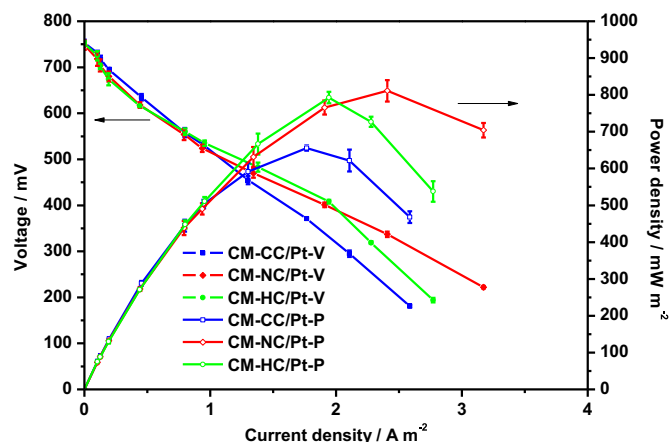


Fig. 2. Polarization curve and power density curve of MFCs with different cathodes.

performances of MFCs using CM-NA and CM-HA anodes exceeded that of the control ($655 \pm 7 \text{ mW m}^{-2}$). The maximum power density of CM-NA and CM-HA anodes was $811 \pm 24 \text{ mW m}^{-2}$ and $777 \pm 35 \text{ mW m}^{-2}$, the corresponding current density was 2.4 A m^{-2} and 1.9 A m^{-2} , respectively. Compared to the control, the maximum power density after modification was enhanced by 24%

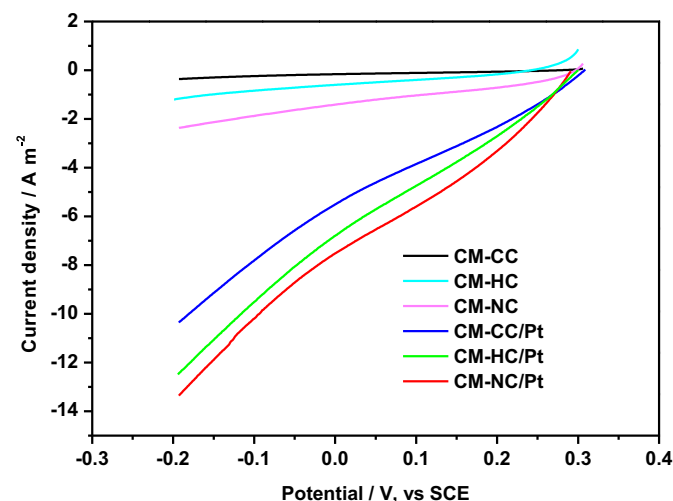


Fig. 1. LSV results (current density vs potential) of modified cathodes before installing in MFCs (50 mM PBS).

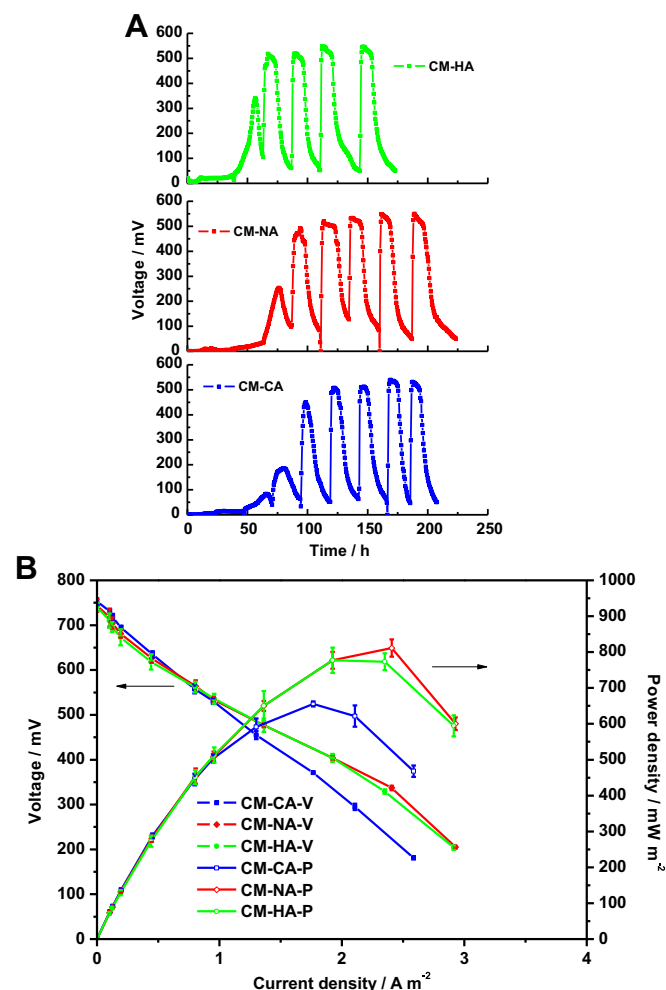


Fig. 3. Performance of MFCs with anodes CM-NA and CM-HA, compared to the control (CM-CA): (A) voltage production, and (B) curves of polarization.

and 19%, respectively. This enhancement in performance, however, was lower than that on the activated carbon fiber felt using the similar modification method (from 1304 mW m^{-2} to 2066 mW m^{-2} , by 58%) [10], which might be due to the difference in anode material [9]. The maximum power density after modification was also comparable with our previous work (792 mW m^{-2}) using the same anode materials but modified by electrochemical oxidation using nitric acid as electrolyte [11].

3.2.2. Surface characteristics of carbon mesh with different treatments

Scanning electron micrographs were taken for the CM-CA, CM-HA, and CM-NA anode before MFCs operation. Fig. 4 illustrates the distinct difference in morphology of these carbon mesh anodes. Compared with the relatively smooth surface of the unmodified anode, the more cracks generated on the modified anodes increased the surface area. Study showed that an increase in the surface area could enhance power density when the materials had the same characteristics [32]. Besides, the changes in the surface chemistry might affect power production [4].

XPS analysis of the different carbon meshes indicated the presence of primarily C, N, and O on the surface, with relatively smaller amounts of F, Si and other elements (Fig. 5). Consistent with the data reported previously [4,33], the magnitude of the N1s/C1s ratio of different treatment methods was correlated with the changes of power density. The N1s/C1s ratio increased from 0.0069 (CM-CA, the control) to 0.0458 (CM-NA) and 0.0221 (CM-HA). In CM-CA, the N1s spectra showed a maximum peak (BE = 401.9 eV) with a shoulder (BE = 400 eV), which were assigned to protonated nitrogen and tertiary amines, respectively [4]. In CM-NA, the atomic ratio of N increased 5.6 times compared to the control (Table 1). The N1s spectra showed a maximum peak (BE = 405.8 eV) with a shoulder (BE = 400.4 eV), which were presumably assigned to ammonium nitrate and pyrrole, accounted for 69.96% and 30.03%, separately [10]. Their contents were contrary to that of activated carbon fiber felt using the same modified method, which might be another reason for lower power [10]. In CM-HA, it showed a maximum peak at BE = 400.9 eV, which was assigned to quaternary nitrogen (i.e. 'graphitic nitrogen') [34]; and a shoulder around 399.7 eV, which was most likely attributed to pyrazoline groups [35]. This change might improve the electrical conductivity of material, and be good for electron transfer. Thus, according to the shortened accumulative time and higher power densities, it could be inferred that the introduction of nitrogen functional groups through modification could favor for bacterial adhesion, and facilitated electron transfer from bacteria to electrodes [6].

XPS results showed that the oxygen content in the spectra changed as a result of different treatments (Table 1). The O1s/C1s

atom ratio of CM-CA was 0.18, which was increased to 0.21 after nitric acid treatment (CM-NA) and decreased to 0.09 after hydrazine hydrate treatment (CM-HA). The relationship between O1s/C1s atom ratios and power densities here showed the same pattern with previous report on anodes modified by acid treatment [4,10]. However, the O1s/C1s atom ratio of CM-HA produced similar results with heat treatment and combined acid-heat treatment anodes in previous study [4]. The XPS spectrum also showed two O1s peaks at 533 eV and 531.7 eV (Fig. 5C), presumably arising from C–O and C=O bonds, respectively. The ratio of C–O to O1s slightly decreased after modification, from 0.93 to 0.91, which was attributed to the increase in power output [4].

3.3. Coupling function of anode and cathode modification

3.3.1. MFC performance

Stable maximum voltages were obtained for all MFCs after inoculation for 120 h. The start-up time of MFCs with hydrazine hydrate-treated anode and cathode (NN) still was the shortest, and that of the nitric acid-treated anode and cathode MFCs (HH) was slightly faster than the unmodified control (CC). Clearly, the modification improved the peak voltage and prolonged the operational cycle (Fig. 6A). Among these MFCs, the highest voltage of HH reactor was 564 mV, which was larger than the unmodified control (532 mV). The period of NN reactor was 32 h, which was 11 h longer than that of the unmodified MFCs (21 h).

Such a longer period and higher output voltage led to the Coulombic efficiency significantly increased from 12.1 to 16.0% (Fig. 6B). The highest CE was obtained from the NN MFCs, increased by 32.2%. And that of HH MFC (14.0%) increased by 16.5%. As observed previously [36], our tests also indicated that the CE increased with current density.

Polarization and power density curves showed that the maximum power densities of NN and HH MFCs were all observed larger than that of the control ($655 \pm 7 \text{ mW m}^{-2}$) (Fig. 6C). The power of HH reactor increased by 38%– $905 \pm 15 \text{ mW m}^{-2}$, which was also larger than CM-NC/Pt MFCs ($811 \pm 29 \text{ mW m}^{-2}$, increased by 24%) (Fig. 2) and CM-NA MFCs ($811 \pm 24 \text{ mW m}^{-2}$, increased by 24%) (Fig. 3B). NN reactor produced the highest power density of $860 \pm 30 \text{ mW m}^{-2}$, which was 31% larger than the control, while it only improved by 21% (cathode modification alone, CM-HC/Pt MFCs) (Fig. 2) and 19% (anode modification alone, CM-HA MFCs) (Fig. 3B).

Fig. 7 shows the cyclic voltammograms of the anodes after bacteria attachment. Compared to the unmodified control, the treated electrodes resulted in larger current responses in potentials ranging between -1 V and 1 V . The much higher electrochemical activity might result from the increased attachment of bacteria due

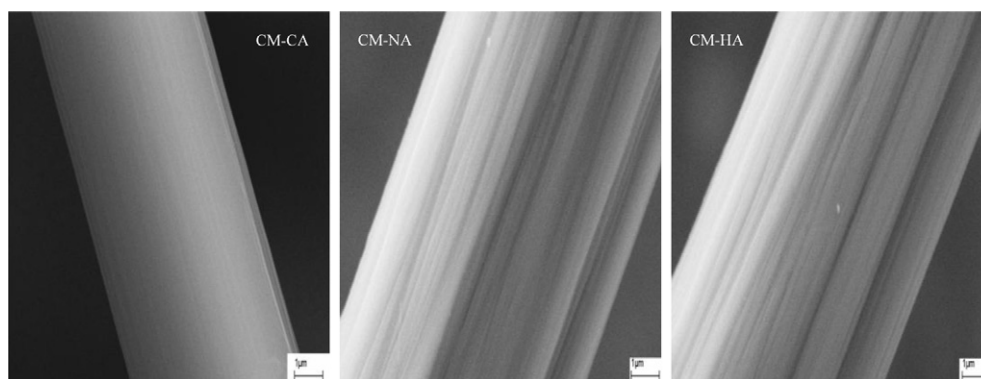


Fig. 4. Electron microscope photograph of unmodified and modified carbon mesh.

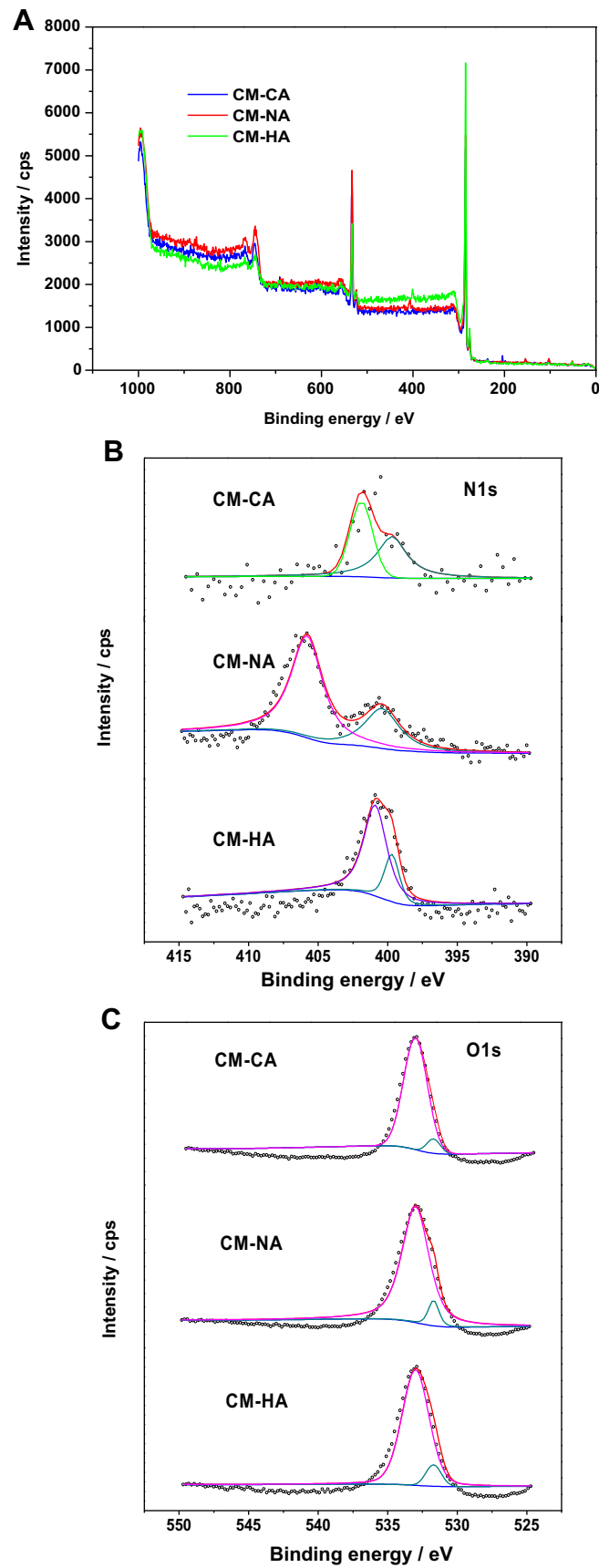


Fig. 5. XPS spectra of the nitric acid and hydrazine hydrate-treated anodes compared to the control: (A) whole spectra, (B) N1s and (C) O1s.

Table 1
Elemental content (%) of the carbon mesh and atomic ratios of N/C, O/C and C–O/O1s with different treatment methods based on the XPS analysis.

Element	CM-CA	CM-NA	CM-HA
C1s (%)	82.14	77.52	88.20
O1s (%)	14.77	16.54	8.35
N1s (%)	0.57	3.55	1.95
N1s/C1s (%)	0.0069	0.0458	0.0221
O1s/C1s (%)	0.18	0.21	0.09
C=O (%)	1.01	1.32	0.75
C–O (%)	13.76	15.22	7.60
C–O/O1s (%)	0.93	0.92	0.91

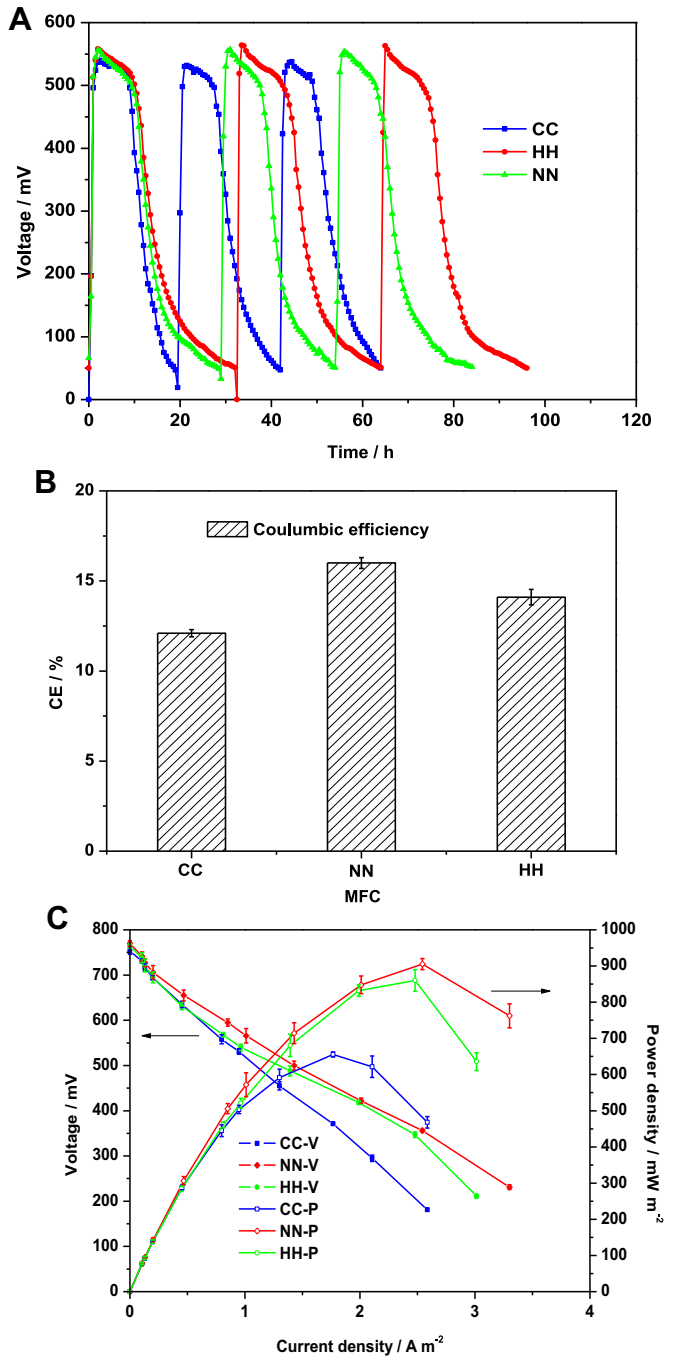


Fig. 6. Performance of MFCs (NN, HH), compared to the control (CC): (A) voltage production, (B) coulombic efficiencies, and (C) power density.

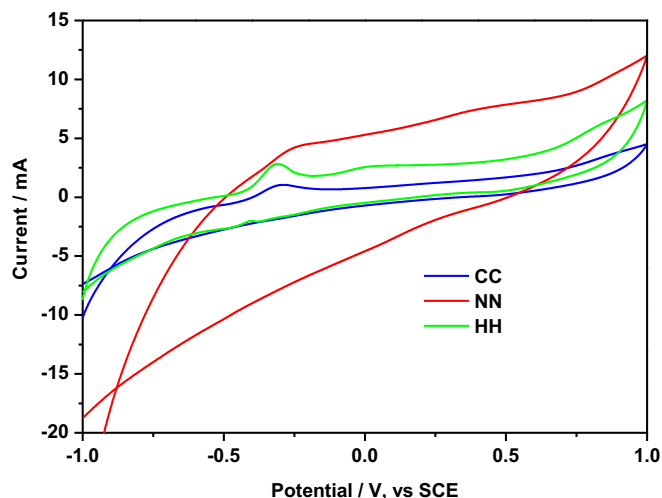


Fig. 7. Cyclic voltammograms of bacteria on different anodes in a PBS solution (50 mM, pH 7.0).

to the enhanced surface area of the treated electrodes [37], or the increased carboxyl containing functional groups on carbon mesh [38]. This fact reflected that the treated anodes had much better electron transfer properties than the unmodified control. The CVs also showed that the bacterial had a redox potential of around -0.3 V against the SCE reference electrode. It was plausible that the electrochemical activity observed here was due to the outer-membrane cytochromes of bacteria [39]. The modification of electrodes enhanced the electron-transfer efficiency from the bacteria to the anode, and consequently increased the peak current.

4. Conclusions

Single-chamber air-cathode MFCs were constructed using chemical modified and unmodified electrodes. In comparison with the unmodified MFCs, chemical modifications of carbon mesh anodes alone including nitric acid treatment and hydrazine hydrate treatment, improved the maximum power densities by 24% and 19%, and shortened the start-up time by 7% and 45%, respectively. The improvement of power generation of the modified anodes might result from changes of surface topography and surface functional groups on carbon mesh, which enabled electrogenic bacteria to adhere and grow better on the anodes.

Additionally, the cathodic modification alone using the two method increased power by 24% and 21% compared to the unmodified control. All modified cathodes showed larger current response as well as higher power density in MFCs. The power density of MFCs was further improved by 38% against the control one when using nitric acid-treated anode and cathode, and indicated the coupling interaction between anode modification and cathode modification. The hydrazine hydrate-treated anode and cathode reactor also improved the maximum power density by 31%. In addition, the average CE values increased from 12.1% to 16.0%. Therefore, the coupling function of anode and cathode modification offered a new method to further increase the performance of MFCs.

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